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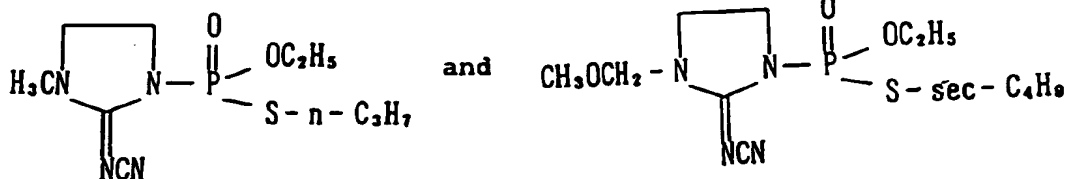
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(54) **Organophosphorus compounds, method for preparing same and insecticides, acaricides and nematocides containing same.**

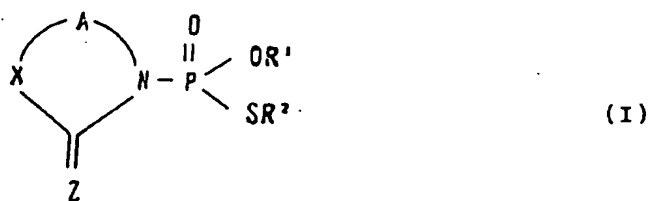
(57) Insecticidal, acaricidal or nematocidal compositions which comprise a series of organophosphorus compounds represented by the compounds represented by the following structural formulae:



show excellent effect for controlling harmful insects, mites and nematodes, do not give off a bad or irritating odor and exhibit very low toxicity to warm-blooded animals.

**EP 0 464 830 A1**

The present invention relates to organophosphorus compounds represented by the following general formula (I):



[ wherein R<sup>1</sup> and R<sup>2</sup> each represents a C<sub>1</sub> to C<sub>4</sub> alkyl group; X represents O, S, CH<sub>2</sub>, CH-R<sup>3</sup> ( wherein R<sup>3</sup> represents a C<sub>1</sub> to C<sub>3</sub> alkyl group), NH or N-R<sup>4</sup> {wherein R<sup>4</sup> represents a C<sub>1</sub> to C<sub>4</sub> alkyl group (the alkyl group may be substituted with at least one group selected from the group consisting of alkoxy, alkylthio, cyano, alkoxyalkoxy and alkylamino groups and halogen atoms), an alkenyl group which may be substituted with halogen atoms, an alkynyl group which may be substituted with halogen atoms, a phosphoric acid ester radical, a cyano group, a group of the following general formula (II):



(wherein R<sup>5</sup> represents an alkyl or alkylamino group which may be substituted with halogen atoms) or a group represented by the following general formula: -(R<sup>6</sup>)<sub>n</sub>-CO-R<sup>7</sup> (wherein n is 0 or 1; R<sup>6</sup> represents a methylene group which may be substituted with alkyl groups, or an ethylene group which may be substituted with alkyl groups; R<sup>7</sup> represents an alkyl group which may be substituted with halogen atoms, an alkoxy group which may be substituted with halogen atoms, an alkylthio group, an alkylamino group or a hydrogen atom); Z represents a group represented by the general formula: N-R<sup>8</sup> (R<sup>8</sup> represents a nitro group, a cyano group, an alkylsulfonyl group which may be substituted with halogen atoms, a tosyl group or an alkylcarbonyl group which may be substituted with halogen atoms) or a group represented by the general formula: C(CN)R<sup>9</sup> (wherein R<sup>9</sup> represents a cyano group or an alkoxy carbonyl group); and A represents an ethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups, a trimethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups or a group represented by the general formula: -CH<sub>2</sub>NR<sup>10</sup>CH<sub>2</sub>- (wherein R<sup>10</sup> is a C<sub>1</sub> to C<sub>3</sub>, alkyl group)]. The compounds of formula (I) according to the present invention specifically exclude organophosphorus compounds of the general formula (I) in which R<sup>1</sup> and R<sup>2</sup> are C<sub>1</sub> to C<sub>4</sub> alkyl groups respectively, X is NH, Z is a cyanoimino group or a nitroimino group and A is an ethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups or a trimethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups. The present invention also relates to a method for preparing the compounds of formula (I), and insecticides, acaricides and nematocides which comprise the compounds as an active ingredient.

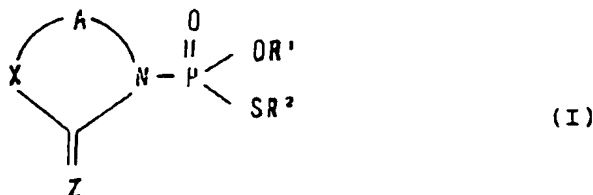
Recently, organophosphorus compounds having an imidazolidinyl group have been investigated and developed. For instance, JP-A- 61-267594 and JP-A- 2-793 disclose that these compounds can be used as insecticides, acaricides, nematocides and agents for killing soil insect pests. However, these patents simply disclose organophosphorus compounds having an imidazolidine skeleton which carries an oxygen or sulfur atom on the 2-position. Moreover, the insecticidal, acaricidal and nematocidal effects of these compounds are insufficient and are not necessarily satisfactory.

Accordingly, an object of the present invention is to provide novel organophosphorus compounds represented by the foregoing general formula (I).

Another object of the present invention is to provide a method for preparing novel organophosphorus compounds represented by the foregoing general formula (I).

A further object of the present invention is to provide an agricultural chemical having insecticidal, acaricidal and nematocidal effects higher than those attained by conventional agricultural chemicals and exhibiting very low toxicity to warm-blooded animals. These objects could be achieved on the basis of the

finding that excellent control of harmful insect pests can be achieved by organophosphorus compounds (hereinafter referred to as "the compounds of the present invention") represented by the following general formula (I):



- 15 [ wherein R<sup>1</sup> and R<sup>2</sup> each represents a C<sub>1</sub> to C<sub>4</sub> alkyl group; X represents O, S, CH<sub>2</sub>, CH-R<sup>3</sup> (wherein R<sup>3</sup> represents a C<sub>1</sub> to C<sub>3</sub> alkyl group), NH or N-R<sup>4</sup> (wherein R<sup>4</sup> represents a C<sub>1</sub> to C<sub>4</sub> alkyl group (the alkyl group may be substituted with at least one group selected from the group consisting of alkoxy, alkylthio, cyano, alkoxyalkyloxy and alkylamino groups and halogen atoms), an alkenyl group which may be substituted with halogen atoms, an alkynyl group which may be substituted with halogen atoms, a phosphoric acid ester radical, a cyano group, a group of the following general formula (II):



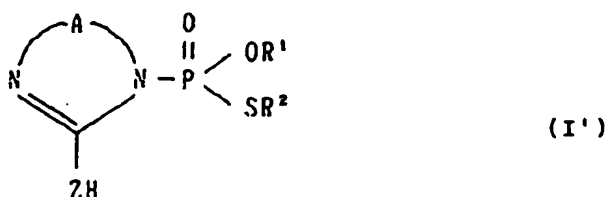
- 25 (wherein R<sup>5</sup> represents an alkyl or alkylamino group which may be substituted with halogen atoms) or a group represented by the following general formula: -(R<sup>6</sup>)<sub>n</sub>-CO-R<sup>7</sup> (wherein n is 0 or 1; R<sup>6</sup> represents a methylene group which may be substituted with alkyl groups, or an ethylene group which may be substituted with alkyl groups; R<sup>7</sup> represents an alkyl group which may be substituted with halogen atoms, an alkoxy group which may be substituted with halogen atoms, an alkylthio group, an alkylamino group or a hydrogen atom); Z represents a group represented by the general formula: N-R<sup>8</sup> (R<sup>8</sup> represents a nitro group, a cyano group, an alkylsulfonyl group which may be substituted with halogen atoms, a tosyl group or an alkylcarbonyl group which may be substituted with halogen atoms) or a group represented by the general formula: C(CN)R<sup>9</sup> (wherein R<sup>9</sup> represents a cyano group or an alkoxy carbonyl group); and A represents an ethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups, a trimethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups or a group represented by the general formula: -CH<sub>2</sub>NR<sup>10</sup>CH<sub>2</sub>- (wherein R<sup>10</sup> is a C<sub>1</sub> to C<sub>3</sub> alkyl group)]. The compounds of formula (I) according to the present invention specifically exclude organophosphorus compounds of the general formula (I) in which R<sup>1</sup> and R<sup>2</sup> are C<sub>1</sub> to C<sub>4</sub> alkyl groups respectively, X is NH, Z is a cyanoimino group or a nitroimino group and A is an ethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups or a trimethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups.

- 45 The terms "alkyl", "alkenyl" and "alkynyl" as used herein alone or as part of another group (e.g. alkoxy, alkylthio, alkylamino, alkylsulfonyl, alkylcarbonyl) include residues having 1 to 6, preferably 1 to 4 carbon atoms - as far as they are not defined otherwise.

These organophosphorus compounds do not give off any bad or irritating odor and have low toxicity to warm-blooded animals. Therefore, they can widely be used and thus have very high usefulness.

- 50 The organophosphorus compounds represented by the foregoing general formula (I) also include stereoisomers such as optical isomers. In addition, in the general formula (I), when X is the group NH, the organophosphorus compounds may exist in the form of a tautomer represented by the following general formula (I'):

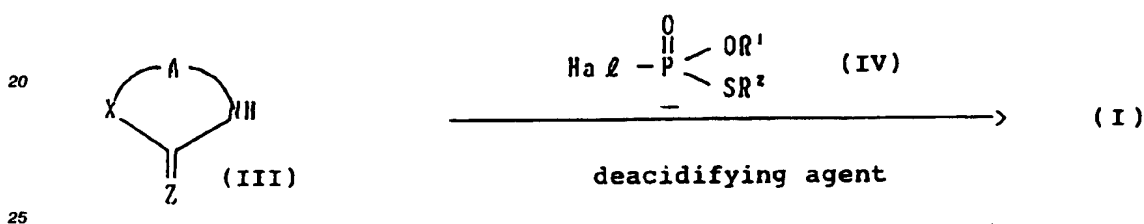
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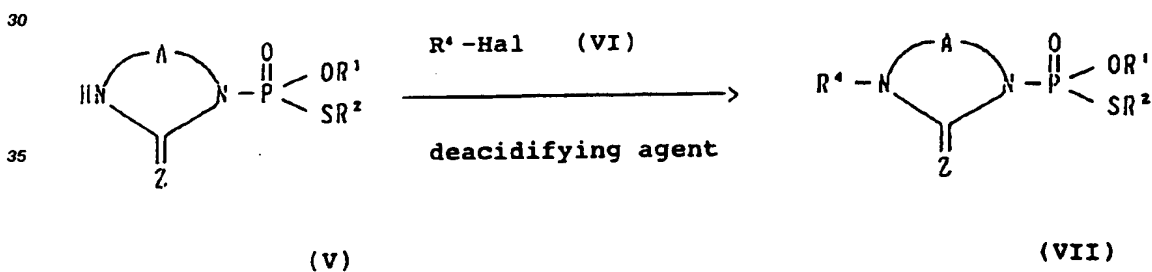
10 (wherein R¹, R², Z and A are the same as those defined above). These compounds (tautomeric isomers) are likewise included in the scope of the compound of the present invention.

The compounds of the present invention can be prepared in accordance with, for instance, any one of the following three methods.

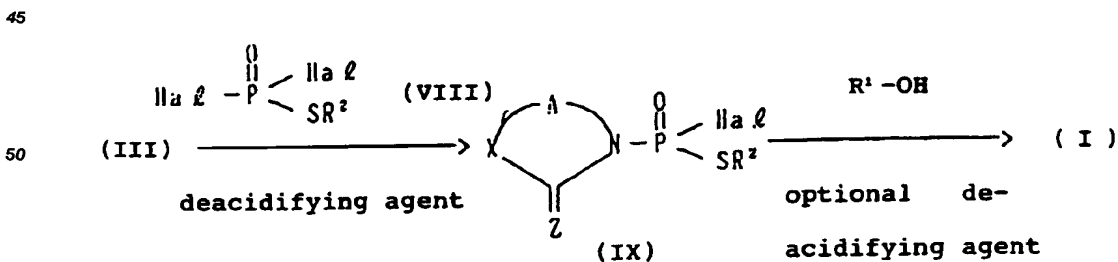
15 Method (i)



Method (ii) (in the formula (I), X is a group: NR⁴)



Method (iii)



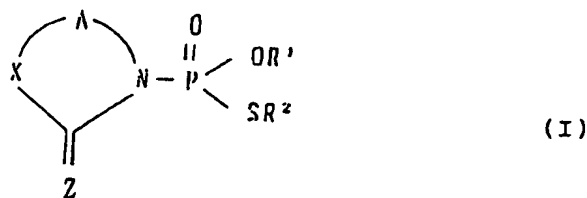
(Wherein R¹, R², R⁴, X, Z and A are the same as those defined above and Hal means a halogen atom).

The foregoing reactions are in general carried out at a temperature ranging from -100 to +60° C and

preferably from  $-80$  to  $+30^{\circ}$  C. Moreover, these reactions are performed in the presence of an acidifying agent, examples of which include organic lithium compounds such as n-butyl lithium, t-butyl lithium and phenyl lithium; inorganic bases such as sodium hydride, potassium hydride, metallic sodium, sodium hydroxide and potassium hydroxide; alkoxides such as sodium methoxide, sodium ethoxide and potassium t-butoxide; and organic bases such as triethylamine and pyridine.

In addition, these reactions are desirably carried out in the presence of a solvent. Examples of such solvents include aromatic hydrocarbons such as benzene, toluene, xylene and chlorobenzene; cyclic or non-cyclic aliphatic hydrocarbons such as hexane and cyclohexane; ethers such as diethyl ether, methyl ethyl ether, dioxane and tetrahydrofuran; nitriles such as acetonitrile, propionitrile and acrylonitrile; and aprotic polar solvents such as dimethylformamide, dimethylsulfoxide, sulfolane and hexamethyl-phosphoric acid triamide.

Typical examples of the compounds of the present invention will now be listed in the following Table 1. Each compound will hereinafter be described by the corresponding number of the compound.



In the following Table 1, the symbol " $\leftarrow$ " as set forth in the column " $\leftarrow$ A—" represents a bond with X and "ph" means a p-phenylene group.

Table 1-1

Comp. No.	R¹	R²	$\leftarrow$ A —	X	Z	Physical Property
1	C₂H₅	n-C₄H₉	-(CH₂)₂-	N-CH₃	N-NO₂	m.p.: 46.0~48.0° C
2	C₂H₅	n-C₄H₉	-(CH₂)₂-	N-C₂H₅	N-NO₂	oily substance
3	C₂H₅	n-C₄H₉	-(CH₂)₂-	N-CH₃	N-CN	oily substance
4	C₂H₅	n-C₄H₉	-(CH₂)₂-	N-C₂H₅	N-CN	oily substance
5	C₂H₅	n-C₄H₉	-(CH₂)₂-	N-n-C₄H₉	N-CN	oily substance
6	C₂H₅	n-C₄H₉	-(CH₂)₂-	N-n-C₄H₉	N-CN	oily substance

7	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -NH	C(CN) <sub>2</sub>	oily substance
8	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -NH	N-SO <sub>2</sub> -ph-4-CH <sub>3</sub>	oily substance
9	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -NH	N-SO <sub>2</sub> CH <sub>3</sub>	oily substance

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Table 1-2

Comp. No.	R <sup>1</sup>	R <sup>2</sup>	-A-	X	Z	Physical Property
10	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	S	N-SO <sub>2</sub> CH <sub>3</sub>	oily substance
11	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	S	N-CN	oily substance
12	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	S	N-NO <sub>2</sub>	oily substance
13	C <sub>6</sub> H <sub>5</sub>	sec-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	oily substance
14	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-P(O)(OC <sub>2</sub> H <sub>5</sub> )-S-n-C <sub>8</sub> H <sub>17</sub>	N-CN	oily substance
15	C <sub>6</sub> H <sub>5</sub>	sec-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH(CH <sub>3</sub> ) <sub>2</sub>	N-CN	oily substance
16	C <sub>6</sub> H <sub>5</sub>	sec-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	m.p.: 45.0~47.0° C
17	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	C(CN) <sub>2</sub>	oily substance
18	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	C(CN) <sub>2</sub>	oily substance
19	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-SO <sub>2</sub> CH <sub>3</sub>	oily substance

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Table 1-3

Comp. No.	R <sup>1</sup>	R <sup>2</sup>	-A-	X	Z	Physical Property
20	C <sub>6</sub> H <sub>5</sub>	sec-C <sub>8</sub> H <sub>17</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	C(CN) <sub>2</sub>	m.p.: 73.0~76.0° C
21	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	CH <sub>3</sub>	-CH <sub>2</sub> NCH <sub>3</sub> -NH	N-NO <sub>2</sub>	oily substance
22	C <sub>6</sub> H <sub>5</sub>	n-C <sub>8</sub> H <sub>17</sub>	ditto	N-CH <sub>3</sub>	N-NO <sub>2</sub>	oi...

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23	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	ditto	N-n-C <sub>3</sub> H <sub>7</sub>	N-NO <sub>2</sub>	oily substance
24	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> CH=CH <sub>2</sub>	N-NO <sub>2</sub>	oily substance
25	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-SO <sub>2</sub> CF <sub>3</sub>	oily substance
26	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> CH=CH <sub>2</sub>	N-CN	oily substance
27	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> C≡CH	N-CN	oily substance
28	C <sub>3</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-COOCH <sub>3</sub>	N-CN	oily substance
29	C <sub>3</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> OCH <sub>3</sub>	N-CN	oily substance

Table 1-4

Comp. No.	R <sup>1</sup>	R <sup>2</sup>	-A-	X	Z	Physical Property
30	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	$\begin{array}{c} \text{CH}_3 \\   \\ -\text{CHCH}_2- \end{array}$	N-CH <sub>2</sub> OC <sub>2</sub> H <sub>5</sub>	N-CN	oily substance
31	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	$\begin{array}{c} \text{CH}_3 \\   \\ -\text{CH}_2\text{CCH}_2- \\   \\ \text{CH}_3 \end{array}$	ditto	N-CN	oily substance
32	C <sub>3</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-SO <sub>2</sub> N(CH <sub>3</sub> ) <sub>2</sub>	N-CN	oily substance
33	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> SC <sub>2</sub> H <sub>5</sub>	N-CN	oily substance
34	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	NOOCF <sub>3</sub>	oily substance
35	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-C <sub>2</sub> H <sub>5</sub>	C(CN) <sub>2</sub>	oily substance
36	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-n-C <sub>3</sub> H <sub>7</sub>	C(CN) <sub>2</sub>	oily substance
37	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	NH	C(CN)COOC <sub>2</sub> H <sub>5</sub>	oily substance
38	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	C(CN)COOC <sub>2</sub> H <sub>5</sub>	oily substance
39	C <sub>3</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-C <sub>2</sub> H <sub>5</sub>	C(CN)COOC <sub>2</sub> H <sub>5</sub>	oily substance

Table 1-5

Comp. No.	R <sup>1</sup>	R <sup>2</sup>	- A -	X	Z	Physical Property
40	C <sub>2</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	CH <sub>3</sub>	N-CN	oily substance
41	CH <sub>3</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	m.p.: 76.0~80.0° C
42	C <sub>2</sub> H <sub>5</sub>	CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	m.p.: 116.0~118.5° C
43	C <sub>2</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	oily substance
44	C <sub>2</sub> H <sub>5</sub>	iso-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	oily substance
			CH <sub>3</sub>			
45	C <sub>2</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-CHCH <sub>2</sub> -	O	N-CN	oily substance
46	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-COOC <sub>2</sub> H <sub>5</sub>	N-CN	oily substance
47	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> CN	N-CN	oily substance
48	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-SO <sub>2</sub> CH <sub>3</sub>	N-CN	oily substance
49	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> OCH <sub>2</sub> CH <sub>2</sub> OCH <sub>3</sub>	N-CN	oily substance
50	C <sub>2</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CON(CH <sub>3</sub> ) <sub>2</sub>	N-CN	oily substance

Table 1-6

Comp. No.	R <sup>1</sup>	R <sup>2</sup>	- A -	X	Z	Physical Property
51	C <sub>2</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-n-C <sub>4</sub> H <sub>9</sub>	N-CN	oily substance
52	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> OCH <sub>3</sub>	N-NO <sub>2</sub>	oily substance
53	C <sub>2</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> OCH <sub>3</sub>	C(CN) <sub>2</sub>	oily substance
54	C <sub>2</sub> H <sub>5</sub>	sec-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CO-C(CH <sub>3</sub> ) <sub>2</sub> -	C(CN) <sub>2</sub>	oily substance
				CH <sub>2</sub> Cl		
55	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>2</sub> CO-	N-CN	oily substance
				C(CH <sub>3</sub> ) <sub>3</sub>		
56	C <sub>2</sub> H <sub>5</sub>	n-C <sub>4</sub> H <sub>9</sub>	-(CH <sub>2</sub> ) <sub>2</sub> -	N-CH <sub>3</sub>	N-CN	m.p.: 58.5~61.5° C



57	C <sub>8</sub> H <sub>8</sub>	n-C <sub>8</sub> H <sub>8</sub>	-(CH <sub>2</sub> ) <sub>7</sub> -CH <sub>3</sub>	N-CN	oily substance
58	C <sub>8</sub> H <sub>8</sub>	n-C <sub>8</sub> H <sub>8</sub>	-(CH <sub>2</sub> ) <sub>6</sub> -CHCH <sub>3</sub>	N-CN	oily substance
59	C <sub>8</sub> H <sub>8</sub>	n-C <sub>8</sub> H <sub>8</sub>	-(CH <sub>2</sub> ) <sub>7</sub> -CH <sub>3</sub>	NNO <sub>2</sub>	oily substance
60	C <sub>8</sub> H <sub>8</sub>	n-C <sub>8</sub> H <sub>8</sub>	-(CH <sub>2</sub> ) <sub>7</sub> -CH <sub>3</sub>	N-CN	m.p.: 44.0-47.0 ° C

Table 2-1

Comp. No.	IR Spectra and NMR Spectra
1	IR (KBr): 1251 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8-2.1 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> CH <sub>2</sub> -CH <sub>2</sub> ), 2.7-3.2 (m, 2H, SCH <sub>2</sub> ), 3.0 (s, 3H, NCH <sub>3</sub> ), 3.8-4.5 (m, 6H, OCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N)
2	IR (neat): 1251 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8-2.1 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> CH <sub>2</sub> -CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> ), 2.7-3.75 (m, 4H, SCH <sub>2</sub> , NCH <sub>2</sub> ), 3.8-4.5 (m, 6H, OCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N)
3	IR (neat): 2176 (C≡N), 1257 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8-2.1 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> CH <sub>2</sub> -CH <sub>2</sub> ), 2.7-3.2 (m, 2H, SCH <sub>2</sub> ), 3.3 (s, 3H, NCH <sub>3</sub> ), 3.5-4.5 (m, 6H, OCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N)

Table 2-2

Comp. No.	IR Spectra and NMR Spectra
4	IR (neat): 2176 (C≡N), 1257 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8-2.1 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> ), 2.7-3.2 (m, 2H, SCH <sub>2</sub> ), 3.5-4.5

(m, 8H, OCH<sub>3</sub>, NCH<sub>2</sub>CH<sub>2</sub>N, NCH<sub>2</sub>)

5 IR (neat): 2170 (C  $\equiv$  N), 1257 (P=O) cm<sup>-1</sup>

5 NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.1 (m, 13H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
CH<sub>2</sub>CH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>), 2.7~3.2 (m, 2H, SCH<sub>2</sub>), 3.4~  
10 4.5 (m, 8H, OCH<sub>3</sub>, NCH<sub>2</sub>CH<sub>2</sub>N, NCH<sub>2</sub>)

6 IR (neat): 2170 (C  $\equiv$  N), 1257 (P=O) cm<sup>-1</sup>

15 NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.1 (m, 15H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
CH<sub>2</sub>CH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>), 2.7~3.2 (m, 2H, SCH<sub>2</sub>),  
3.5~4.5 (m, 8H, SCH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>N, NCH<sub>2</sub>)

Table 2-3

Comp. No. IR Spectra and NMR Spectra

25 7 IR (neat): 3232 (NH), 2194 (C $\equiv$  N), 1257 (P=O) cm<sup>-1</sup>  
NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.1 (m, 8H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
30 CH<sub>2</sub>CH<sub>2</sub>), 2.7~3.2 (m, 2H, SCH<sub>2</sub>), 3.5~4.5 (m, 6H, OCH<sub>3</sub>,  
NCH<sub>2</sub>CH<sub>2</sub>N), 8.00 (bs, 1H, NH)

8 IR (neat): 3376 (NH), 1257 (P=O) cm<sup>-1</sup>  
35 NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.1 (m, 8H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>CH<sub>2</sub>  
CH<sub>2</sub>), 2.36 (s, 3H, Ph-CH<sub>3</sub>), 2.6~3.1 (m, 2H, SCH<sub>2</sub>),  
40 3.4~4.4 (m, 6H, OCH<sub>3</sub>, NCH<sub>2</sub>CH<sub>2</sub>N), 7.1~7.9 (m, 5H, NH,  
benzene)

9 IR (neat): 3370 (NH), 1257 (P=O) cm<sup>-1</sup>  
45 NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.1 (m, 8H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
CH<sub>2</sub>CH<sub>2</sub>), 2.7~3.2 (m, 2H, SCH<sub>2</sub>), 2.93 (s, 3H,  
50 SO<sub>2</sub>CH<sub>3</sub>), 3.5~4.5 (m, 6H, OCH<sub>3</sub>, NCH<sub>2</sub>CH<sub>2</sub>N), 7.34  
(bs, 1H, NH)

Table 2-4

Comp. N .	IR Spectra and NMR Spectra
10	IR (neat): 1257 (P=O) $\text{cm}^{-1}$ NMR ( $\text{CDCl}_3$ ), $\delta$ (ppm): 0.8~2.1 (m, 8H, $\text{OCH}_2\text{CH}_2$ , $\text{SCH}_2\text{-CH}_2\text{CH}_2$ ), 2.7~3.5 (m, 4H, $\text{SCH}_2$ , $\text{SCH}_2\text{CH}_2\text{N}$ ), 2.99 (s, 3H, $\text{SO}_2\text{CH}_3$ ), 3.9~4.5 (m, 4H, $\text{OCH}_2$ , $\text{SCH}_2\text{CH}_2\text{N}$ )
11	IR (neat): 2176 ( $\text{C} \equiv \text{N}$ ), 1257 (P=O) $\text{cm}^{-1}$ NMR ( $\text{CDCl}_3$ ), $\delta$ (ppm): 0.8~2.1 (m, 8H, $\text{OCH}_2\text{CH}_2$ , $\text{SCH}_2\text{CH}_2$ ), 2.7~4.7 (m, 8H, $\text{SCH}_2$ , $\text{SCH}_2\text{CH}_2\text{N}$ , $\text{OCH}_2$ )
12	IR (neat): 1251 (P=O) $\text{cm}^{-1}$ NMR ( $\text{CDCl}_3$ ), $\delta$ (ppm): 0.8~2.1 (m, 8H, $\text{OCH}_2\text{CH}_2$ , $\text{SCH}_2\text{-CH}_2\text{CH}_2$ ), 2.7~3.4 (m, 4H, $\text{SCH}_2$ , $\text{SCH}_2\text{CH}_2\text{N}$ ), 4.0~4.6 (m, 4H, $\text{OCH}_2$ , $\text{SCH}_2\text{CH}_2\text{N}$ )

Table 2-5

Comp. No.	IR Spectra and NMR Spectra
13	IR (neat): 2176 ( $\text{C} \equiv \text{N}$ ), 1257 (P=O) $\text{cm}^{-1}$ NMR ( $\text{CDCl}_3$ ), $\delta$ (ppm): 0.8~2.2 (m, 13H, $\text{OCH}_2\text{CH}_2$ , $\text{SCH-CH}_2\text{CH}_2\text{CH}_2$ , $\text{NCH}_2\text{CH}_2\text{CH}_2\text{N}$ ), 3.1~4.5 (m, 7H, $\text{SCH}$ , $\text{OCH}_2$ , $\text{NCH}_2\text{CH}_2\text{CH}_2\text{N}$ ), 3.35 (s, 3H, $\text{NCH}_3$ )
14	IR (neat): 2182 ( $\text{C} \equiv \text{N}$ ), 1266 (P=O) $\text{cm}^{-1}$ NMR ( $\text{CDCl}_3$ ), $\delta$ (ppm): 0.8~2.1 (m, 16H, $\text{OCH}_2\text{CH}_2$ , $\text{OCH}_2\text{-CH}_2$ , $\text{SCH}_2\text{CH}_2\text{CH}_2$ , $\text{SCH}_2\text{CH}_2\text{CH}_2$ ), 2.6~3.2 (m, 4H, $\text{SCH}_2$ , $\text{SCH}_2$ ), 3.9~4.5 (m, 8H, $\text{OCH}_2$ , $\text{OCH}_2$ , $\text{NCH}_2\text{CH}_2\text{N}$ )
15	IR (neat): 2176 ( $\text{C} \equiv \text{N}$ ), 1257 (P=O) $\text{cm}^{-1}$

NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.0 (m, 17H, OCH<sub>2</sub>CH<sub>2</sub>, SCH-(CH<sub>2</sub>)CH<sub>2</sub>CH<sub>2</sub>, N-CH(CH<sub>2</sub>)<sub>2</sub>), 3.2~4.5 (m, 7H, SCH, NCH<sub>2</sub>CH<sub>2</sub>N, OCH<sub>2</sub>), 4.7~5.2 (m, 1H, NCH)

Table 2-6

Comp. No.	IR Spectra and NMR Spectra
16	IR (neat): 2176 (C $\equiv$ N), 1254 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> ), 3.20~4.5 (m, 7H, SCH, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 3.32 (s, 3H, NCH <sub>3</sub> )
17	IR (neat): 2200 (C $\equiv$ N), 1263 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-CH <sub>2</sub> CH <sub>2</sub> ), 2.1~3.7 (m, 8H, NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> N, SCH <sub>2</sub> ), 3.35 (s, 3H, NCH <sub>3</sub> ), 3.9~4.5 (m, 2H, OCH <sub>2</sub> )
18	IR (neat): 2200 (C $\equiv$ N), 1269 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-CH <sub>2</sub> CH <sub>2</sub> ), 2.6~4.5 (m, 8H, SCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 3.30 (s, 3H, NCH <sub>3</sub> )

Table 2-7

Comp. No.	IR Spectra and NMR Spectra
19	IR (neat): 1617 (C = N)cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-CH <sub>2</sub> CH <sub>2</sub> ), 2.6~4.5 (m, 8H, SCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 2.97 (s, 3H, SO <sub>2</sub> CH <sub>3</sub> ), 3.30 (s, 3H, NCH <sub>3</sub> )

Table 2-8

Comp. No.	IR Spectra and NMR Spectra
20	IR (KBr): 2200 (C≡N), 1265 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> ), 3.20~4.5 (m, 7H, SCH, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 3.30 (s, 3H, NCH <sub>3</sub> )
21	IR (neat): 3274 (NH), 1248 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> ), 2.56 (s, 3H, NCH <sub>3</sub> ), 2.70~3.20 (m, 2H, SCH <sub>2</sub> ), 3.9~4.7 (m, 6H, OCH <sub>2</sub> , NCH <sub>2</sub> N, NCH <sub>2</sub> N), 9.49 (bs, 1H, NH)
22	IR (neat): 1248 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> ), 2.58 (s, 3H, NCH <sub>3</sub> ), 3.0 (s, 3H, NCH <sub>3</sub> ), 2.70~3.2 (m, 2H, SCH <sub>2</sub> ), 3.8~4.5 (m, 6H, NCH <sub>2</sub> N, NCH <sub>2</sub> N)

Table 2-9

Comp. No.	IR Spectra and NMR Spectra
24	IR (neat): 1572 (C=N), 1251 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> ), 2.7~3.2 (m, 2H, SCH <sub>2</sub> ), 3.7~4.5 (m, 8H, OCH <sub>2</sub> , NCH <sub>2</sub> CH=CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N), 5.1~6.0 (m, 3H, NCH <sub>2</sub> CH=CH <sub>2</sub> )
25	IR (neat): 1515 (C=N), 1248 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -

$\text{CH}_2 \text{CH}_2$ ), 2.7~3.2 (m, 2H,  $\text{SCH}_2$ ), 3.13 (s, 3H,  $\text{NCH}_2$ )  
 3.6~4.5 (m, 6H,  $\text{OCH}_2$ ,  $\text{NCH}_2\text{CH}_2\text{N}$ )

IR (neat): 2176 ( $\text{C} \equiv \text{N}$ ), 1257 ( $\text{P}=\text{O}$ )  $\text{cm}^{-1}$

NMR ( $\text{CDCl}_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 8H,  $\text{OCH}_2\text{CH}_2$ ,  $\text{SCH}_2$ -  
 $\text{CH}_2 \text{CH}_2$ ), 2.7~3.2 (m, 2H,  $\text{SCH}_2$ ), 3.8~4.5 (m, 8H,  
 $\text{OCH}_2$ ,  $\text{NCH}_2\text{CH}=\text{CH}_2$ ,  $\text{NCH}_2\text{CH}_2\text{N}$ ), 5.1~6.0 (m, 3H,  
 $\text{NCH}_2\text{CH}=\text{CH}_2$ )

Table 2-10

Comp. No.

## IR Spectra and NMR Spectra

IR (neat): 3232 ( $\text{C} \equiv \text{CH}$ ), 2182 ( $\text{C} \equiv \text{N}$ ), 1257 ( $\text{P}=\text{O}$ )  $\text{cm}^{-1}$

NMR ( $\text{CDCl}_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 8H,  $\text{OCH}_2\text{CH}_2$ ,  $\text{SCH}_2$ -  
 $\text{CH}_2 \text{CH}_2$ ), 2.38 (t, 1H,  $\text{CH}_2\text{C} \equiv \text{CH}$ ), 2.7~3.2 (m, 2H,  
 $\text{SCH}_2$ ), 3.6~4.5 (m, 6H,  $\text{OCH}_2$ ,  $\text{NCH}_2\text{CH}_2\text{N}$ ), 5.55 (d,  
 2H,  $\text{NCH}_2\text{C} \equiv \text{CH}$ )

IR (neat): 2188 ( $\text{C} \equiv \text{N}$ ), 1737 ( $\text{C}=\text{O}$ ), 1257 ( $\text{P}=\text{O}$ )  $\text{cm}^{-1}$

NMR ( $\text{CDCl}_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 11H,  $\text{OCH}_2\text{CH}_2$ ,  $\text{SCH}$ -  
 $(\text{CH}_2)\text{CH}_2 \text{CH}_2$ ), 2.54 (s, 3H,  $\text{COCH}_3$ ), 3.2~4.7 (m, 7H,  
 $\text{SCH}$ ,  $\text{NCH}_2\text{CH}_2\text{N}$ ,  $\text{OCH}_2$ )

IR (neat): 2182 ( $\text{C} \equiv \text{N}$ ), 1257 ( $\text{P}=\text{O}$ )  $\text{cm}^{-1}$

NMR ( $\text{CDCl}_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 11H,  $\text{OCH}_2\text{CH}_2$ ,  $\text{SCH}$ -  
 $(\text{CH}_2)\text{CH}_2 \text{CH}_2$ ), 3.2~4.5 (m, 7H,  $\text{SCH}$ ,  $\text{NCH}_2\text{CH}_2\text{N}$ ,  $\text{OCH}_2$ ),  
 3.41 (s, 3H,  $\text{CH}_3\text{O}$ ), 5.11 (s, 2H,  $\text{NCH}_2\text{O}$ )

Table 2-11

Comp. No.

## IR Spectra and NMR Spectra

- 30 IR (neat): 2182 ( $C \equiv N$ ), 1260 ( $P=O$ )  $cm^{-1}$   
 NMR ( $CDCl_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 14H,  $OCH_2CH_2$ ,  $SCH_2-$   
 5  $CH_2CH_2$ ,  $OCH_2CH_2$ ,  $NCH(CH_2)CH_2N$ ), 2.7~4.5 (m, 9H,  
 $SCH_2$ ,  $OCH_2$ ,  $OCH_2$ ,  $NCH(CH_2)CH_2N$ ), 5.14 (s, 2H,  
 10  $NCH_2O$ )
- 31 IR (neat): 2176 ( $C \equiv N$ ), 1272 ( $P=O$ )  $cm^{-1}$   
 NMR ( $CDCl_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 17H,  $OCH_2CH_2$ ,  $SCH_2-$   
 15  $CH_2CH_2$ ,  $OCH_2CH_2$ ,  $NCH_2C(CH_2)_2CH_2N$ ), 2.6~4.5 (m,  
 10H,  $SCH_2$ ,  $OCH_2$ ,  $OCH_2$ ,  $NCH_2C(CH_2)_2CH_2N$ ), 5.16 (s,  
 20 2H,  $NCH_2O$ )
- 32 IR (neat): 2188 ( $C \equiv N$ ), 1263 ( $P=O$ )  $cm^{-1}$   
 NMR ( $CDCl_3$ ),  $\delta$  (ppm): 0.8~2.0 (m, 11H,  $OCH_2CH_2$ ,  $SCH-$   
 25  $(CH_2)CH_2CH_2$ ), 2.95 (s, 6H,  $SO_2N(CH_2)_2$ ), 3.2~4.5  
 (m, 7H,  $SCH$ ,  $NCH_2CH_2N$ ,  $OCH_2$ )

Table 2-12

Comp. No.	IR Spectra and NMR Spectra
33	IR (neat): 2176 ( $C \equiv N$ ), 1266 ( $P=O$ ) $cm^{-1}$ NMR ( $CDCl_3$ ), $\delta$ (ppm): 0.8~2.0 (m, 8H, $OCH_2CH_2$ , $SCH_2-$ 40 $CH_2CH_2$ ), 2.25 (s, 3H, $SCH_3$ ), 2.7~3.2 (m, 2H, $SCH_2$ ), 3.6~4.5 (m, 6H, $NCH_2CH_2N$ , $OCH_2$ ), 4.90 (s, 2H, $SCH_2N$ )
34	IR (neat): 1662 ( $C=O$ ), 1599 ( $C=N$ ), 1257 ( $P=O$ ) $cm^{-1}$ NMR ( $CDCl_3$ ), $\delta$ (ppm): 0.8~2.0 (m, 8H, $OCH_2CH_2$ , $SCH_2-$ 45 $CH_2CH_2$ ), 2.6~3.2 (m, 2H, $SCH_2$ ), 2.91 (s, 3H, 50 $NCH_3$ ), 3.6~4.5 (m, 6H, $NCH_2CH_2N$ , $OCH_2$ )

35 IR (neat): 2200 (C  $\equiv$  N), 1263 (P=O) cm<sup>-1</sup>  
 NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8-2.0 (m, 11H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
 5 CH<sub>2</sub>CH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>), 2.6-3.2 (m, 2H, SCH<sub>2</sub>), 3.5-  
 4.5 (m, 8H, NCH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>N, OCH<sub>2</sub>)

Table 2-13

Comp. No.	IR Spectra and NMR Spectra
36	IR (neat): 2200 (C $\equiv$ N), 1265 (P=O) cm <sup>-1</sup> 15 NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8-2.0 (m, 13H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - CH <sub>2</sub> CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> ), 2.6-3.2 (m, 2H, SCH <sub>2</sub> ), 20 3.5-4.5 (m, 8H, NCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> )
37	IR (neat): 3292 (NH), 2200 (C $\equiv$ N), 1671 (C=O), 1254 (P=O) cm <sup>-1</sup> 25 NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8-2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - CH <sub>2</sub> CH <sub>2</sub> , COOCH <sub>2</sub> CH <sub>2</sub> ), 2.6-3.2 (m, 2H, SCH <sub>2</sub> ), 3.5- 4.5 (m, 8H, NCH <sub>2</sub> CH <sub>2</sub> N, COOCH <sub>2</sub> CH <sub>2</sub> , OCH <sub>2</sub> ), 9.43 (bs, 30 1H, NH)
38	IR (neat): 2188 (C $\equiv$ N), 1686 (C=O), 1257 (P=O) cm <sup>-1</sup> 35 NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8-2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - CH <sub>2</sub> CH <sub>2</sub> , COOCH <sub>2</sub> CH <sub>2</sub> ), 2.6-3.2 (m, 2H, SCH <sub>2</sub> ), 3.15 (s, 3H, NCH <sub>3</sub> ), 3.4-4.5 (m, 8H, NCH <sub>2</sub> CH <sub>2</sub> N, COOCH <sub>2</sub> - 40 CH <sub>2</sub> , OCH <sub>2</sub> )

Table 2-14

Comp. No.	IR Spectra and NMR Spectra
39	IR (neat): 2188 (C $\equiv$ N), 1686 (C=O), 1257 (P=O) cm <sup>-1</sup>



NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.0 (m, 14H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
CH<sub>2</sub>CH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>, COOCH<sub>2</sub>CH<sub>2</sub>), 2.6~3.2 (m, 2H,  
SCH<sub>2</sub>), 3.4~4.5 (m, 10H, NCH<sub>2</sub>CH<sub>2</sub>N, NCH<sub>2</sub>, COOCH<sub>2</sub>CH<sub>2</sub>,  
OCH<sub>2</sub>)

IR (neat): 2188 (C $\equiv$ N), 1608 (C=N), 1254 (P=O) cm<sup>-1</sup>  
NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.3 (m, 13H, OCH<sub>2</sub>CH<sub>2</sub>, SCH-  
(CH<sub>2</sub>)CH<sub>2</sub>CH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>C=NCN), 2.8~4.4 (m, 7H,  
NCH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>C=NCN, SCH, OCH<sub>2</sub>)

IR (KBr): 2176 (C $\equiv$ N), 1254 (P=O) cm<sup>-1</sup>  
NMR (CDCl<sub>3</sub>),  $\delta$  (ppm): 0.8~2.0 (m, 8H, SCH(CH<sub>2</sub>)CH<sub>2</sub>CH<sub>2</sub>),  
3.32 (s, 3H, NCH<sub>2</sub>), 3.2~3.9 (m, 5H, SCH, NCH<sub>2</sub>CH<sub>2</sub>N),  
3.81 (d, 3H, OCH<sub>2</sub>)

Table 2-15

Comp. No.	IR Spectra and NMR Spectra
42	IR (KBr): 2176 (C $\equiv$ N), 1254 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 1.35 (t, 3H, OCH <sub>2</sub> CH <sub>2</sub> ), 2.40 (d, 3H, SCH <sub>2</sub> ), 3.32 (s, 3H, NCH <sub>2</sub> ), 3.6~4.5 (m, 6H, N- CH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> )
43	IR (neat): 2176 (C $\equiv$ N), 1254 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 1.35 (t, 6H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> CH <sub>2</sub> ), 2.6~3.2 (m, 2H, SCH <sub>2</sub> ), 3.32 (s, 3H, NCH <sub>2</sub> ), 3.6~4.5 (m, 6H, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> )
44	IR (neat): 2176 (C $\equiv$ N), 1260 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.9~2.1 (m, 10H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - CH(CH <sub>2</sub> )), 2.7~3.1 (m, 2H, SCH <sub>2</sub> ), 3.32 (s, 3H, NCH <sub>2</sub> ), 3.6~4.5 (m, 6H, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> )

Table 2-16

Comp. No.	IR Spectra and NMR Spectra
45	IR (neat): 2212 (C $\equiv$ N), 1260 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 14H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> , OCH(CH <sub>2</sub> )CH <sub>2</sub> N), 3.2~4.5 (m, 5H, SCH, OCH(CH <sub>2</sub> )CH <sub>2</sub> N, OCH <sub>2</sub> ), 4.8~5.4 (m, 1H, OCH(CH <sub>2</sub> )CH <sub>2</sub> N)
46	IR (neat): 2194 (C $\equiv$ N), 1764 (C=O)cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> , COOCH <sub>2</sub> CH <sub>2</sub> ), 2.72~3.26 (m, 2H, SCH <sub>2</sub> ), 3.8~4.5 (m, 8H, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> , COOCH <sub>2</sub> CH <sub>2</sub> )
47	IR (neat): 2182 (C $\equiv$ N), 1260 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> ), 2.70~3.22 (m, 2H, SCH <sub>2</sub> ), 3.6~4.5 (m, 6H, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 4.62 (s, 2H, NCH <sub>2</sub> CN)

Table 2-17

Comp. No.	IR Spectra and NMR Spectra
48	IR (neat): 2188 (C $\equiv$ N), 1260 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> ), 2.6~3.2 (m, 2H, SCH <sub>2</sub> ), 3.45 (s, 3H, SO <sub>2</sub> CH <sub>3</sub> ), 3.6~4.5 (m, 6H, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> )
49	IR (neat): 2182 (C $\equiv$ N), 1257 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -CH <sub>2</sub> CH <sub>2</sub> ), 2.7~3.2 (m, 2H, SCH <sub>2</sub> ), 3.35 (s, 3H, OCH <sub>3</sub> ), 3.4~4.5 (m, 10H, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> CH <sub>2</sub> O, OCH <sub>2</sub> CH <sub>3</sub> ), 5.20

(s, 2H, OCH<sub>2</sub>N)

5

Table 2-18

Comp. No.	IR Spectra and NMR Spectra
50	IR (neat): 2182 (C $\equiv$ N), 1710 (C=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> ), 3.05 (s, 6H, N(CH <sub>3</sub> ) <sub>2</sub> ), 3.2~4.5 (m, 7H, SCH, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> )
51	IR (neat): 2176 (C $\equiv$ N), 1254 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 16H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> ), 3.2~4.5 (m, 9H, SCH, NCH <sub>2</sub> CH <sub>2</sub> N, NCH <sub>2</sub> , OCH <sub>2</sub> )
52	IR (neat): 1563 (C=N), 1248 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 8H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-CH <sub>2</sub> CH <sub>2</sub> ), 2.7~4.5 (m, 8H, SCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 3.32 (s, 3H, OCH <sub>3</sub> ), 4.70 (s, 2H, OCH <sub>2</sub> N)

30

Table 2-19

Comp. No.	IR Spectra and NMR Spectra
53	IR (neat): 2206 (C $\equiv$ N), 1560 (C=C) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 11H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> ), 3.2~4.5 (m, 7H, SCH, NCH <sub>2</sub> CH <sub>2</sub> N, OCH <sub>2</sub> ), 3.39 (s, 3H, OCH <sub>3</sub> ), 5.01 (s, 2H, OCH <sub>2</sub> N)
54	IR (neat): 2212 (C $\equiv$ N), 1719 (C=O), 1563 (C=C) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), $\delta$ (ppm): 0.8~2.0 (m, 17H, OCH <sub>2</sub> CH <sub>2</sub> , SCH-(CH <sub>2</sub> )CH <sub>2</sub> CH <sub>2</sub> , NCOC(CH <sub>3</sub> ) <sub>2</sub> ), 3.2~3.8 (m, 3H, SCH,

50

55

CH<sub>2</sub>Cl), 4.0~4.5 (m, 6H, OCH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>N)

55 IR (neat): 2176 (C ≡ N), 1722 (C=O), 1617 (C=N) cm<sup>-1</sup>

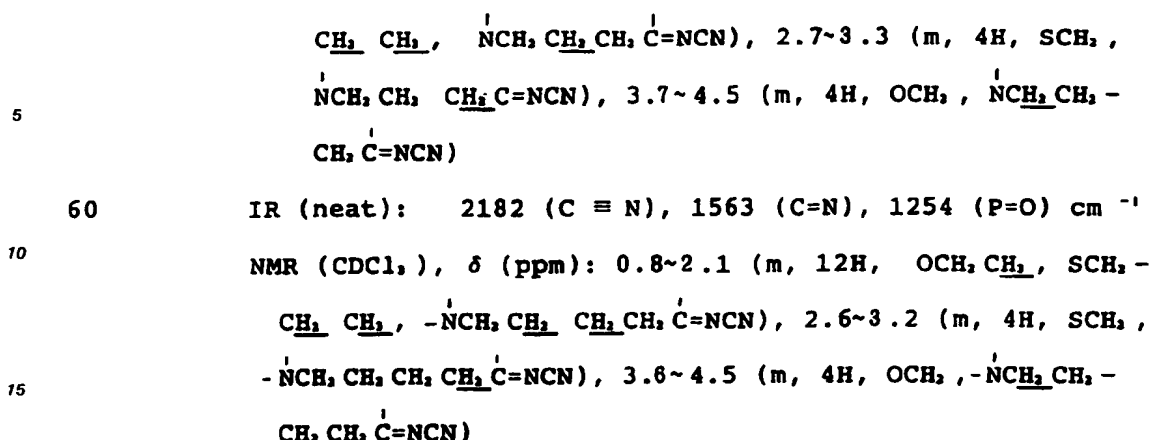
5 NMR (CDCl<sub>3</sub>), δ (ppm): 0.8~2.1 (m, 8H, OCH<sub>2</sub>CH<sub>2</sub>, SCH<sub>2</sub>-  
CH<sub>2</sub>CH<sub>2</sub>), 1.21 (s, 9H, C(CH<sub>3</sub>)<sub>3</sub>), 2.7~4.8 (m, 10H,  
10 OCH<sub>2</sub>, SCH<sub>2</sub>, NCH<sub>2</sub>CH<sub>2</sub>N, NCH<sub>2</sub>CO)

Table 2-20

Comp. No.	IR Spectra and NMR Spectra
56	IR (neat): 2170 (C ≡ N), 1590 (C=N) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.2 (m, 10H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - CH <sub>2</sub> CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> N), 2.6~4.5 (m, 8H, SCH <sub>2</sub> , OCH <sub>2</sub> , 20 NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> N), 3.37 (s, 3H, NCH <sub>3</sub> )
57	IR (neat): 2188 (C ≡ N), 1608 (C=N), 1257 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.4 (m, 10H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - 25 CH <sub>2</sub> CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> C≡NCN), 2.7~3.2 (m, 4H, SCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> C≡NCN), 3.8~4.5 (m, 4H, OCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> - 30 CH <sub>2</sub> C≡NCN)
58	IR (neat): 2188 (C ≡ N), 1605 (C=N), 1257 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~3.4 (m, 16H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> - 35 CH <sub>2</sub> CH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> (CH <sub>2</sub> )C≡NCN), 3.7~4.5 (m, 4H, OCH <sub>2</sub> , NCH <sub>2</sub> CH <sub>2</sub> CH <sub>2</sub> C≡NCN)

Table 2-21

Comp. No.	IR Spectra and NMR Spectra
59	IR (neat): 1542 (C=N), 1263 (P=O) cm <sup>-1</sup> NMR (CDCl <sub>3</sub> ), δ (ppm): 0.8~2.4 (m, 10H, OCH <sub>2</sub> CH <sub>2</sub> , SCH <sub>2</sub> -



The compounds of the present invention exhibit excellent activity as active ingredients of insecticides, acaricides and nematocides. The compounds of the present invention are effective for controlling, for instance, agricultural and horticultural pests such as Coleoptera (for instance, Scarabaeidae, Chrysomelidae, Henosepilachna vigintioctopunctata and Lissorhoptrus oryzophilus), Lepidoptera (for instance, Mamestra brassicae, Pieris rapae, Plutera xylostella, Noctuidae, Adoxophyes orana and Chilo suppressalis), Hemiptera (for instance, Delphacidae, Deltocephalidae, Aleyrodidae, Aphididae and Scales) and Thysanoptera (for instance, Scirtothrips dorsalis and Thrips pulmi); Sanitary pests such as Mosquitoes, Flies, Blattidae, Fleas and Lice; Grain pests; cloth and house pests; Plant parasitic nematodes such as root-knot nematodes and root-lesion nematodes; and Plant parasitic mites such as Tetranychus urticae, Tetranychus cinnabarinus, Tetranychus kanzawai and Panonychus citri. They are also effective for controlling soil insect pests. The term "soil insect pest" used herein means, for instance, Gastropoda such as slugs and snails; Isopoda such as Armadillidium vulgare and Sow bugs. In addition, they are likewise effective for controlling insect pest such as Dicotyl, organophosphorus compound-resistant plant parasitic mites, organophosphorus compound-resistant Aphididae and Musca domestica.

When the compounds of the present invention are used as active ingredients of insecticidal, acaricidal, nematocidal compositions and composition for killing or controlling soil pest insect, they may be used as such, but may in general be formed into a variety of formulations together with adjuvants such as emulsifiable concentrates, dusts, wettable powders, liquid formulations, aerosols and pastes like conventional formulations of agricultural chemicals. The formulations in general comprise 0.5 to 90 parts by weight of the active ingredient and 10 to 99.5 parts by weight of an adjuvant. When these formulations are practically used, they can be applied as such or after diluting them with a diluent such as water to a desired concentration.

The term "adjuvant" herein means carriers, emulsifying agents, suspending agents, dispersing agents, spreaders, penetrants, wetting agents, thickeners and stabilisers. These adjuvants may, if necessary, be added in an appropriate amount. The carriers can roughly be classified into solid carriers and liquid carriers. Examples of the solid carriers include vegetable and animal powder such as starches, active carbon, soybean flour, wheat flour, wood powder, fish meal and powdered milk; and mineral powders such as talc, kaolin, bentonite, calcium carbonate, zeolite, diatomaceous earth, white carbon, clay, alumina and powdered sulfur. On the other hand, examples of the liquid carriers are water; alcohols such as methyl alcohol and ethylene glycol; ketones such as acetone and methyl ethyl ketone; ethers such as dioxane and tetrahydrofuran; aliphatic hydrocarbons such as kerosine; aromatic hydrocarbons such as xylene, trimethylbenzene, tetramethylbenzene, cyclohexane and solvent naphtha; halogenated hydrocarbons such as chloroform and chlorobenzene; amides such as dimethylformamide; esters such as ethyl acetate and glycerin esters of fatty acids; nitriles such as acetonitrile; and sulfur atom-containing compounds such as dimethylsulfoxide. If necessary, the compounds of the present invention may be admixed with or used simultaneously with other agricultural chemicals such as insecticides, acaricides, nematocides, bactericides, antiviral agents, attractants, herbicides and plant growth regulators. In admixtures, greater effectiveness may sometimes be attained.

Examples of insecticides, acaricides or nematocides include organophosphorus acid ester compounds such as DDVP, Diazinon, Malathion, Fenitrothion, Prothiofos, Dioxabenzofos and Acephate; carbamate

compounds such as Carbaryl, Propoxur, Oxamyl, Carbofuran and M thomyl; organochlorin compounds such as Dicofol and Tetradifon; organometal compounds such as Cyhexatin and Fenbutatin oxide; pyrethroid compounds such as Fenvalerat, Permethrin, Deltamethrin and Bifenthrin; urea compounds such as Diflubenzuron, Thiflubenuron and Chlorfluazuron; heterocyclic compounds such as Buprofezin and Hexythiazox; and other compounds such as dinitro compounds, organosulfur compounds, amidine compounds and triazine compounds. In addition to the foregoing compounds, the compound of the present invention can also be admixed with or used simultaneously with microbial pesticides such as BT agents and insect pathogen virus pesticides.

Examples of the bactericides are organophosphorus compounds such as Iprobenfos, Edifenphos and Phosethyl-aluminum; organocopper compounds such as oxyquinoline copper, copper terephthalate; organochlorine compounds such as Fthalide and Chlorothalonil; dithiocarbamate compounds such as Maneb, Zineb and Propineb; dicarboximide compounds such as Iprodione, Vinclozolin and Procymidone; azole compounds such as Triadimefon, Bitertanol, Etaconazole, Propiconazole and Penconazole; benzimidazole compounds such as Thiophanate-methyl and Benomyl; carbinol compounds such as Fenarimol and Flutriafol; benzanilide compounds such as Mepronil and Flutolanil; phenylamide compounds such as Metalaxyl and Oxadixyl; and other compounds such as piperazine compounds, quinoxaline compounds, morpholine compounds, anthraquinone compounds, sulfenic acid compounds, crotonic acid compounds, urea compounds and antibiotics.

As has been discussed above, the insecticides, acaricides and nematocides which comprise, as active ingredient, the compounds of the present invention are effective for controlling a variety of deleterious insects, harmful mites, destructive nematodes and harmful soil insect pests. These agricultural chemicals or formulations are applied at a concentration ranging from 1 to 20,000 ppm, desirably 20 to 2,000 ppm expressed in terms of the amount of the active ingredient. The concentration of these active ingredients can be appropriately adjusted depending on various factors such as shapes of the formulations, methods, purposes, time and places of application as well as the condition of infestation of insect pest. For instance, when aquatic insect pests are to be controlled, the insect pest can be controlled by spraying a solution of the compound having a concentration falling within the range defined above and, therefore, the concentration of the active ingredient required for controlling the aquatic insect pest is lower than that defined above. The amount of the agricultural chemicals or formulations to be applied per unit area (per 10 a) ranges from about 0.1 to 5,000 g and preferably 10 to 1,000 g expressed in terms of the amount of the active ingredient, but in a particular case, they may be applied in an amount outside the range defined above.

The application of a variety of formulations or dilute solutions thereof which comprise the compounds of the present invention can be performed according to any manner of application usually adopted, for instance, spraying (such as spraying, dusting, misting, atomizing, granule application and application on water surface); soil applications (such as mixing and drench); surface applications (such as coating, dressing and painting); and poison bait. It is also possible to control the development and growth of harmful insect pests, in particular deleterious insects through the action of excreta obtained by admixing the foregoing active ingredient to feed and giving the feed to domestic animals. Further, they can also be applied in accordance with ultra low volume application. In this method, the chemicals or formulations can comprise 100% active ingredient.

#### (EXAMPLES)

The method for preparing the compounds of the present invention will hereinafter be explained in more detail with reference to the following Preparation Examples.

Preparation Example 1: O-ethyl-S-n-propyl-(3-ethyl-2-nitroimino-1-imidazolidinyl)phosphonothiolate (Compound No. 2)

2.00 g of O-ethyl-S-n-propyl-(2-nitroimino-1-imidazolidinyl) phosphonothiolate was dissolved in 20 ml of N,N-dimethylformamide and then 0.32 g of a 60% sodium hydride was gradually added to the resulting solution. Thereafter, the solution was cooled down to 0 to 5 °C and 1.27 g of ethyl iodide was gradually added thereto dropwise. After the completion of the dropwise addition, the temperature of the solution was slowly brought back to room temperature and the solution was further stirred for additional 12 hours to complete the reaction. After the reaction, the solution was poured into ice water and extracted with dichloromethane. The dichloromethane phase was washed with water, dried over anhydrous magnesium sulfate, the dichloromethane was distilled off under reduced pressure and the residue obtained was purified by silica gel column chromatography (eluent: ethyl acetate) to give 0.50 g of Compound No. 2 as an oily

substance.

Preparation Example 2: O-ethyl-S-n-propyl-(3-methyl-2-cyanoimino-1-imidazolidinyl)phosphonothiolate (Compound No. 3)

5 To a mixture comprising 0.44 g of a 60% sodium hydride and 50 ml of N,N-dimethylformamide, 1.24 g of 1-methyl-2-cyanoiminoimidazolidine was gradually added. After the mixture was allowed to stand for a brief period, 2.76 g of an 88.2% toluene solution of O-ethyl-S-n-propylphosphorochloride thiolate was gradually dropwise added to the mixture. After the completion of the dropwise addition, the reaction solution  
10 was poured into ice water and then extracted with chloroform. The chloroform phase was washed with water, dried over anhydrous magnesium sulfate, the chloroform was distilled off under reduced pressure and the residue obtained was purified by silica gel column chromatography (eluent: chloroform/methanol = 97:3) to give 1.50 g of Compound No. 3 as an oily substance.

15 Preparation Example 3: O-ethyl-S-n-propyl-(2-methanesulfonylimino-1-imidazolidinyl)phosphonothiolate (Compound No. 9)

2.0 g of 2-methanesulfonyliminoimidazolidine was dissolved in 20 ml of N,N-dimethylformamide. The resulting solution was cooled down to 0° C and then 1.02 g of a 60% sodium hydride was gradually added.  
20 Thereafter, 3.00 g of an 88.2% toluene solution of O-ethyl-S-n-propylphosphorochloride thiolate was gradually dropwise added to the solution. After the completion of the dropwise addition, the temperature of the reaction solution was slowly brought back to room temperature and further the reaction was continued for additional 12 hours. After the reaction, the reaction solution was poured into ice water, neutralized with a 10% hydrochloric acid solution and then extracted with chloroform. The chloroform phase was washed with  
25 water, dried over anhydrous magnesium sulfate, the chloroform was distilled off under reduced pressure and the residue obtained was purified by silica gel column chromatography (eluent: chloroform/methanol = 97:3) to give 2.20 g of Compound No. 9 as an oily substance.

30 Preparation Example 4: O-ethyl-S-sec-butyl-(3-methoxymethyl-2-cyanoimino-1-imidazolidinyl)-phosphonothiolate (Compound No. 29)

1.50 g of O-ethyl-S-sec-butyl-(2-cyanoimino-1-imidazolidinyl) phosphonothiolate was dissolved in 20 ml of tetrahydrofuran. The resulting solution was cooled and maintained at a temperature of ranging from 0 to 5° C and 0.25 g of a 60% sodium hydride was gradually added to the solution. After 30 minutes, 0.50 g of  
35 chloromethyl methyl ether was dropwise added thereto. After the completion of the dropwise addition, the temperature of the reaction solution was brought back to room temperature and the solution was stirred for additional 3 hours to complete the reaction.

After the reaction, the reaction solution was poured into ice water and then extracted with ethyl acetate. The ethyl acetate phase was washed with water, dried over anhydrous magnesium sulfate, the ethyl acetate  
40 was distilled off under reduced pressure and the residue obtained was purified by silica gel column chromatography (eluent: chloroform) to thus give 1.10 g of Compound No. 29 as an oily substance.

Specific examples of formulations will be described below, but the adjuvants such as carriers and surfactants are by no means limited to those used in the following examples. In the following specific formulations, the term "part" means "part by weight" unless otherwise specified.

45 Formulation Example 1 (Wettable Powder)

20 parts of Compound No. 1, 56 parts of acid clay, 15 parts of white carbon, 4 parts of calcium lignin sulfonate and 5 parts of polyoxyethylene alkylphenyl ether were uniformly mixed and pulverized to give a  
50 wettable powder.

Formulation Example 2 (Emulsifiable Concentrate)

To 20 parts of Compound No. 2, 75 parts of xylene was added. And then, 5 parts of New Calgen ST-20  
55 (available from Takemoto Oil & Fat Co., Ltd.) was added to the resulting solution as an emulsifying agent and then the mixture was mixed and dissolved to give an emulsifiable concentrate.

Formulation Example 3 (Granule)

5 parts of Compound No. 1, 3 parts of calcium lignin sulfonate, one part of sodium dodecylbenzenesulfonate, 30 parts of bentonite and 61 parts of clay were sufficiently pulverized and mixed. The mixture was then sufficiently kneaded while adding water, followed by granulation and drying to thus give a granule.

The insecticidal, acaricidal and nematocidal effects of the compounds of the present invention will hereunder be explained with reference to the following Test Examples.

#### Test Example 1

The emulsifiable concentrate obtained by Formulation Example 2 was diluted with water to 500 ppm and 50 ppm and the resulting diluted emulsions were sprayed on leaves of chinese cabbage. After air-drying, the leaves were introduced into a plastic container having a size of 21 cm (height)X 13 cm (width) x 3 cm (depth), then 10 third instar larvae of common cutworm were put on the leaves. The container was then placed in a thermostated room maintained at 26° C. After 2 days, the number of surviving larvae was recorded to obtain the mortality (%) (repeated two times). The results thus obtained are summarized in the following Table 3.

Table 3

<u>Compound No.</u>	<u>Concn. of Active Ingredient (ppm)</u>	<u>Mortality (%)</u>
2	500	100
	50	0
3	500	100
	50	70
4	500	100
	50	70
5	500	100
	50	90
6	500	100
	50	100
7	500	100
	50	100
9	500	100



EP 0 464 830 A1

		50	80
	10	500	100
5		50	0
	14	500	100
		50	100
10	15	500	100
		50	90
	16	500	100
15		50	20
	17	500	100
		50	60
20	18	500	100
		50	100
25	20	500	100
		50	90
	21	500	100
30		50	0
	26	500	100
		50	10
35	27	500	100
		50	0
	29	500	100
40		50	20
	30	500	100
45		50	100
	31	500	100

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**EP 0 464 830 A1**

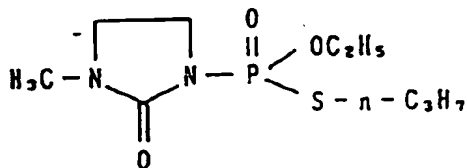
		50	50
5	32	500	100
		50	10
	33	500	100
10		50	70
	34	500	100
		50	100
15	35	500	100
		50	80
	36	500	100
20		50	20
	37	500	100
		50	90
25	38	500	100
		50	50
30	39	500	100
		50	40
	40	500	100
35		50	0
	41	500	100
		50	100
40	44	500	100
		50	90
	45	500	100
45		50	90
	46	500	100

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		50	100
5	47	500	100
		50	50
	49	500	100
10		50	0
	50	500	100
		50	10
15	51	500	100
		50	50
20	53	500	100
		50	10
25	Comparative Compound A	500	100
		50	10
	Untreated	---	0

The Comparative Compound A as set forth in Table 3 is a compound represented by the following structural formula and disclosed in Japanese Unexamined Patent Publication No. Sho 61-267594:



#### Test Example 2

The emulsifiable concentrate obtained by Formulation Example 2 was diluted with water to 500 ppm and the seedlings of rice plant were immersed therein for 10 seconds. After air-drying, the seedlings whose roots were wrapped round with absorbent cotton were introduced into a test tube, then 10 second instar larvae of green rice leafhopper were put into the test tube, the opening thereof was closed with gauze. The tube was placed in a thermostated room maintained at 26 °C. After 2 days, the number of surviving larvae was recorded to obtain the mortality (%) (repeated two times). The results thus obtained are summarized in the following Table 4.

Table 4

	<u>Compound N .</u>	<u>C ncn. of Active Ingredient (ppm)</u>	<u>Mortality (%)</u>
5	1	500	100
	3	500	100
10	4	500	100
	5	500	100
	9	500	100
15	10	500	100
	11	500	100
20	14	500	100
	15	500	100
	16	500	100
25	17	500	100
	18	500	100
30	19	500	100
	20	500	100
35	21	500	100
	26	500	100
	27	500	100
40			
45			
50			
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EP 0 464 830 A1

	28	500	100
	29	500	100
5	30	500	100
	31	500	100
10	32	500	100
	33	500	100
	34	500	100
15	35	500	100
	37	500	100
20	38	500	100
	40	500	100
	41	500	100
25	44	500	100
	45	500	100
30	46	500	100
	47	500	100
35	49	500	100
	50	500	100
	51	500	100
40	53	500	100
	Comparative Compound A	500	100
45	Untreated	---	0

The Comparative Compound A as set forth in Table 4 is the same as that defined above in connection with Table 3.

50 Test Example 3

A kidney bean leaf was cut into 3 x 5 cm<sup>2</sup>. The leaf was put on a filter paper wetted with water for preventing drying of the leaf and 20 female adults of kanzawa spider mite were released on the leaf. After 24 hours, 500 ppm and 50 ppm of concentration of active ingredient prepared by diluting with water the emulsifiable concentrate obtained by Formulation Example 2 were sprayed on the leaf. After air-drying, the mites were held in a thermostated room maintained at 26 ° C. After 24 hours, the number of surviving adults were recorded to obtain the mortality (%) (repeated two times). The results thus obtained are summarized in the following Table 5.

Table 5

	<u>Compound No.</u>	<u>Concn. of Active Ingr dient (ppm)</u>	<u>Mortality (%)</u>
5	1	500	100
		50	0
10	2	500	100
		50	0
	3	500	100
15		50	80
	4	500	100
20		50	20
	5	500	100
		50	40
25	6	500	100
		50	50
30	7	500	100
35			
40			
45			
50			
55			

**EP 0 464 830 A1**

		50	90
	9	500	100
5		50	60
	10	500	100
10		50	70
	11	500	100
		50	80
15	12	500	100
		50	0
	14	500	100
20		50	80
	15	500	100
		50	10
25	16	500	100
		50	0
	17	500	100
30		50	30
	18	500	100
		50	90
35	19	500	100
		50	0
40	20	500	100
		50	0
	26	500	100
45		50	30
	27	500	100

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EP 0 464 830 A1

		50	20
	28	500	100
5		50	30
	29	500	100
		50	50
10	30	500	100
		50	20
	31	500	100
15		50	10
	32	500	100
		50	0
20	33	500	100
		50	0
25	34	500	100
		50	20
	35	500	100
30		50	10
	36	500	100
		50	10
35	37	500	100
		50	50
	38	500	100
40		50	20
	39	500	100
		50	0
45	40	500	100

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EP 0 464 830 A1

		50	10
	41	500	100
5		50	80
	44	500	100
		50	10
10	45	500	100
		50	10
	46	500	100
15		50	20
	47	500	100
		50	20
20	50	500	100
		50	0
25	51	500	100
		50	10
	53	500	100
30		50	0
	Comparative Compound A	500	100
		50	10
35	Untreated	---	0

The Comparative Compound A is the same as that defined above in connection with Table 3.

40 Test Example 4

To a 1/5,000 are pot, soil contaminated with root-knot nematodes was packed and the soil was admixed with the granules obtained by Formulation Example 3 in an amount of 250 g and 25 g/are expressed in terms of the amount of the active ingredient. Two days after the treatment, seedlings of tomato at 3rd to 45 4th-leaf stage were transplanted in the soil. Twenty-five days after the transplantation, the degree of the invasion of root-knot nematodes (root-knot index) was determined (repeated two times).

The root-knot index was evaluated on the basis of the following evaluation criteria:

- 0 no invasion of root-knot nematodes
- 1 slight invasion of root-knot nematodes
- 50 2 medium invasion of root-knot nematodes
- 3 severe invasion of root-knot nematodes
- 4 very severe invasion of root-knot nematodes

The results thus obtained are summarized in the following Table 6.

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Table 6

5	Compound	Amount of Active Ingredient	<u>Root-Knot Index</u>
	<u>No.</u>	<u>(g/are)</u>	
	1	250	0
		25	3
10	2	250	0
		25	2
15			
20			
25			
30			
35			
40			
45			
50			
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EP 0 464 830 A1

	3	250	0
		25	0
5	4	250	0
		25	0
10	5	250	0
		25	1
	6	250	0
15		25	3
	7	250	0
		25	1
20	8	250	0
		25	2
25	9	250	0
		25	0
30	10	250	0
		25	2
	12	250	0
35		25	3
	14	250	0
		25	0
40	15	250	0
		25	0
45	16	250	0
		25	1
50	17	250	0
		25	0

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EP 0 464 830 A1

	18	250	0
		25	0
5	20	250	0
		25	0
10	21	250	0
		25	3
	25	250	0
15		25	3
	26	250	0
		25	0
20	27	250	0
		25	0
25	29	250	0
		25	0
30	30	250	0
		25	0
	31	250	0
35		25	0
	33	250	0
		25	2
40	34	250	0
		25	0
45	35	250	0
		25	0
50	36	250	0
		25	2

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**EP 0 464 830 A1**

	37	250	0
		25	0
5	38	250	0
		25	0
10	39	250	0
		25	0
	40	250	0
15		25	0
	41	250	0
		25	0
20	43	250	0
		25	3
	44	250	0
25		25	0
	45	250	0
		25	3
30	47	250	0
		25	3
35	49	250	0
		25	2
	50	250	0
40		25	0
	51	250	0
		25	0
45	53	250	0
		25	0

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	54	250	0
		25	1
5	Comparative Compound A	250	0
		25	2
	Comparative Compound B	250	0
10		25	1
	Untreated	---	4

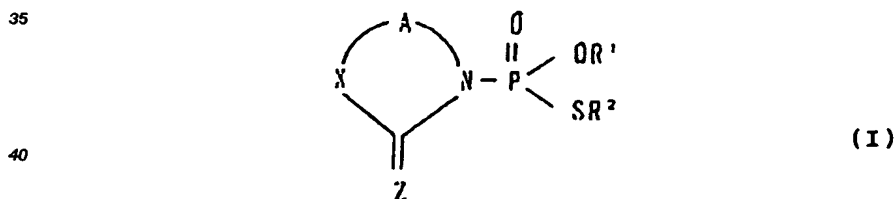
The Comparative Compound A as set forth in Table 6 is the same as that defined above in connection with Table 3 and Comparative Compound B is a compound (oxamyl) represented by the following structural formula:



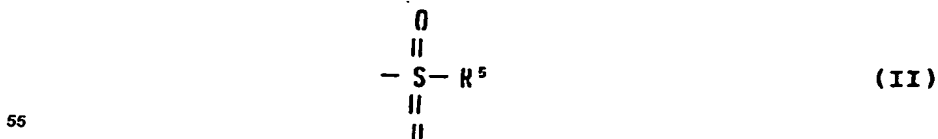
The compounds of the present invention represented by the foregoing general formula (I) exhibit effectiveness in controlling harmful insects, mites and nematodes and thus can be used as novel insecticides, acaricides and nematocides which do not give off a bad or irritating odor and have low toxicity to warm-blooded animals.

### Claims

1. An organophosphorus compound represented by the following general formula (I):



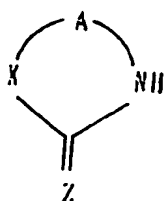
[ wherein  $\text{R}^1$  and  $\text{R}^2$  each represents a  $\text{C}_1$  to  $\text{C}_4$  alkyl group; X represents O, S,  $\text{CH}_2$ ,  $\text{CH-R}^3$  (wherein  $\text{R}^3$  represents a  $\text{C}_1$  to  $\text{C}_3$  alkyl group), NH or  $\text{N-R}^4$  { wherein  $\text{R}^4$  represents a  $\text{C}_1$  to  $\text{C}_4$  alkyl group (the alkyl group may be substituted with at least one group selected from the group consisting of alkoxy, alkylthio, cyano, alkoxyalkoxy and alkylamino groups and halogen atoms), an alkenyl group which may be substituted with halogen atoms, an alkynyl group which may be substituted with halogen atoms, a phosphoric acid ester radical, a cyano group, a group of the following general formula (II):



(wherein  $\text{R}^5$  represents an alkyl or alkylamino group which may be substituted with halogen atoms) or a

group represented by the following general formula:  $-(R^6)_n-CO-R^7$  (wherein  $n$  is 0 or 1;  $R^6$  represents a methylene group which may be substituted with alkyl groups, or an ethylene group which may be substituted with alkyl groups;  $R^7$  represents an alkyl group which may be substituted with halogen atoms, an alkoxy group which may be substituted with halogen atoms, an alkylthio group, an alkylamino group or a hydrogen atom);  $Z$  represents a group represented by the general formula:  $N-R^8$  ( $R^8$  represents a nitro group, a cyano group, an alkylsulfonyl group which may be substituted with halogen atoms, a tosyl group or an alkylcarbonyl group which may be substituted with halogen atoms) or a group represented by the general formula:  $C(CN)R^9$  (wherein  $R^9$  represents a cyano group or an alkoxy carbonyl group); and  $A$  represents an ethylene group which may be substituted with  $C_1$  to  $C_3$  alkyl groups, a trimethylene group which may be substituted with  $C_1$  to  $C_3$  alkyl groups or a group represented by the general formula:  $-CH_2NR^{10}-CH_2-$  (wherein  $R^{10}$  is a  $C_1$  to  $C_3$  alkyl group); and wherein formula (I) according to the present invention specifically excludes organophosphorus compounds of the general formula (I) in which  $R^1$  and  $R^2$  are  $C_1$  to  $C_4$  alkyl groups,  $X$  is  $NH$ ,  $Z$  is a cyanoimino group or a nitroimino group and  $A$  is an ethylene group which may be substituted with  $C_1$  to  $C_3$  alkyl groups or a trimethylene group which may be substituted with  $C_1$  to  $C_3$  alkyl groups.

2. The organophosphorus compound according to claim 1 which is O-ethyl-S-n-propyl-(3-methyl-2-cyanoimino-1-imidazolidinyl) phosphonothiolate.
3. The organophosphorus compound according to claim 1 which is O-ethyl-S-n-propyl-(3-ethyl-2-cyanoimino-1-imidazolidinyl) phosphonothiolate.
4. The organophosphorus compound according to claim 1 which is O-ethyl-S-n-propyl-(2-dicyanomethylene-1-imidazolidinyl) phosphonothiolate.
5. The organophosphorus compound according to claim 1 which is O-ethyl-S-n-propyl-(2-methanesulfonylimino-1-imidazolidinyl) phosphonothiolate.
6. The organophosphorus compound according to claim 1 which is O-ethyl-S-sec-butyl-(3-methyl-2-cyanoimino-tetrahydropyrimidinyl) phosphonothiolate.
7. The organophosphorus compound according to claim 1 which is O-ethyl-S-sec-butyl-(3-methyl-2-cyanoimino-1-imidazolidinyl) phosphonothiolate.
8. The organophosphorus compound according to claim 1 which is O-ethyl-S-n-propyl-(3-methyl-2-dicyanomethylene-1-imidazolidinyl) phosphonothiolate.
9. The organophosphorus compound according to claim 1 which is O-ethyl-S-sec-butyl-(3-methyl-2-dicyanomethylene-1-imidazolidinyl) phosphonothiolate.
10. The organophosphorus compound according to claim 1 which is O-ethyl-S-sec-butyl-(3-methoxymethyl-2-cyanoimino-1-imidazolidinyl) phosphonothiolate.
11. The organophosphorus compound according to claim 1 which is O-ethyl-S-n-propyl-[2-(2-cyano-2-ethoxycarbonylmethylene)-1-imidazolidinyl]-phosphonothiolate.
12. The organophosphorus compound according to claim 1 which is O-ethyl-S-sec-butyl-1-pyrrolidinyl-phosphonothiolate.
13. A method for preparing an organophosphorus compound as set forth in claim 1 comprising reacting, in the presence of a deacidifying agent, a heterocyclic compound represented by the following general formula (III):



(III)

[ wherein X represents O, S, CH<sub>2</sub>, CH-R<sup>3</sup> (wherein R<sup>3</sup> represents a C<sub>1</sub> to C<sub>3</sub> alkyl group), NH or N-R<sup>4</sup> { wherein R<sup>4</sup> represents a C<sub>1</sub> to C<sub>4</sub> alkyl group (the alkyl group may be substituted with at least one group selected from the group consisting of alkoxy, alkylthio, cyano, alkoxyalkyloxy and alkylamino groups and halogen atoms), an alkenyl group which may be substituted with halogen atoms, an alkynyl group which may be substituted with halogen atoms, a phosphoric acid ester radical, a cyano group, a group of the following general formula (II):

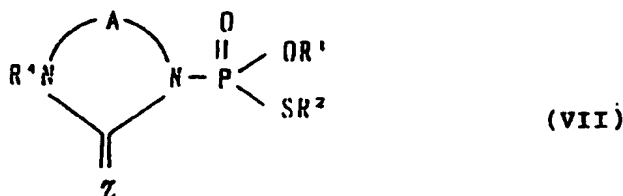


(wherein R<sup>5</sup> represents an alkyl or alkylamino group which may be substituted with halogen atoms) or a group represented by the following general formula: -(R<sup>6</sup>)<sub>n</sub>-CO-R<sup>7</sup> (wherein n is 0 or 1; R<sup>6</sup> represents a methylene group which may be substituted with alkyl groups, or an ethylene group which may be substituted with alkyl groups; R<sup>7</sup> represents an alkyl group which may be substituted with halogen atoms, an alkoxy group which may be substituted with halogen atoms, an alkylthio group, an alkylamino group or a hydrogen atom); Z represents a group represented by the general formula: N-R<sup>8</sup> (R<sup>8</sup> represents a nitro group, a cyano group, an alkylsulfonyl group which may be substituted with halogen atoms, a tosyl group or an alkylcarbonyl group which may be substituted with halogen atoms) or a group represented by the general formula: C(CN)R<sup>9</sup> (wherein R<sup>9</sup> represents a cyano group or an alkoxy carbonyl group); and A represents an ethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups, a trimethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups or a group represented by the general formula: -CH<sub>2</sub>NR<sup>10</sup>CH<sub>2</sub>- (wherein R<sup>10</sup> represents a C<sub>1</sub> to C<sub>3</sub> alkyl group) ], with a phosphoric acid compound represented by the following general formula (IV):



(wherein R<sup>1</sup> and R<sup>2</sup> each represents a C<sub>1</sub> to C<sub>4</sub> alkyl group and Hal represents a halogen atom).

14. A method for preparing an organophosphorus compound represented by the following general formula (VII):

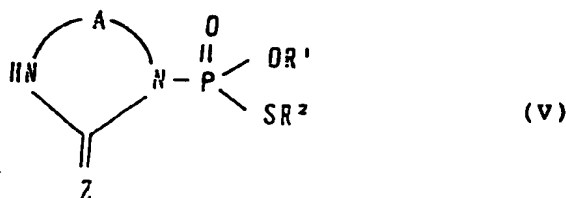




[ wherein R<sup>1</sup> and R<sup>2</sup> each represents a C<sub>1</sub> to C<sub>4</sub> alkyl group; R<sup>4</sup> represents a C<sub>1</sub> to C<sub>4</sub> alkyl group (the alkyl group may be substituted with at least one group selected from the group consisting of alkoxy, alkylthio, cyano, alkoxyalkoxy and alkylamino groups and halogen atoms), an alkenyl group which may be substituted with halogen atoms, an alkynyl group which may be substituted with halogen atoms, a phosphoric acid ester radical, a cyano group, a group of the following general formula (II):



(wherein R<sup>5</sup> represents an alkyl or alkylamino group which may be substituted with halogen atoms) or a group represented by the following general formula: -(R<sup>6</sup>)<sub>n</sub>-CO-R<sup>7</sup> (wherein n is 0 or 1; R<sup>6</sup> represents a methylene group which may be substituted with alkyl groups, or an ethylene group which may be substituted with alkyl groups; R<sup>7</sup> represents an alkyl group which may be substituted with halogen atoms, an alkoxy group which may be substituted with halogen atoms, an alkylthio group, an alkylamino group or a hydrogen atom); Z represents a group represented by the general formula: N-R<sup>8</sup> (R<sup>8</sup> represents a nitro group, a cyano group, an alkylsulfonyl group which may be substituted with halogen atoms, a tosyl group or an alkylcarbonyl group which may be substituted with halogen atoms) or a group represented by the general formula: C(CN)R<sup>9</sup> (wherein R<sup>9</sup> represents a cyano group or an alkoxy carbonyl group); and A represents an ethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups, a trimethylene group which may be substituted with C<sub>1</sub> to C<sub>3</sub> alkyl groups or a group represented by the general formula: -CH<sub>2</sub>NR<sup>10</sup>-CH<sub>2</sub>- (wherein R<sup>10</sup> is a C<sub>1</sub> to C<sub>3</sub> alkyl group) comprising reacting, in the presence of a deacidifying agent, a compound represented by the following general formula (V):

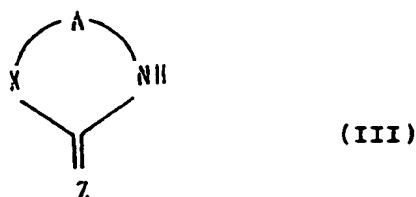


(wherein R<sup>1</sup>, R<sup>2</sup>, A and Z are the same as those defined above) with a compound represented by the general formula (VI):



(wherein R<sup>4</sup> is the same as that defined above; and Hal represents a halogen atom).

15. A method for preparing an organophosphorus compound as set forth in claim 1 comprising reacting, in the presence of a deacidifying agent, a heterocyclic compound represented by the following general formula (III):

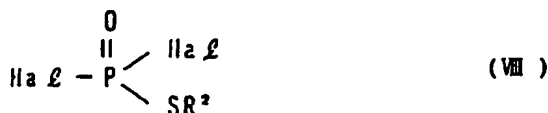


[ wherein X represents O, S, CH<sub>2</sub>, CH-R<sup>3</sup> (wherein R<sup>3</sup> represents a C<sub>1</sub> to C<sub>3</sub> alkyl group), NH or N-R<sup>4</sup> {

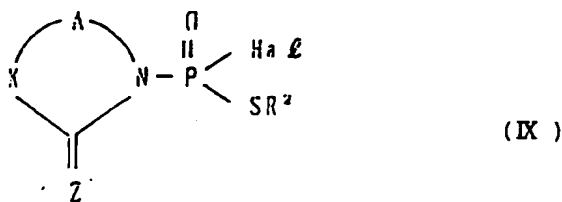
wherein  $R^4$  represents a  $C_1$  to  $C_4$  alkyl group (the alkyl group may be substituted with at least one group selected from the group consisting of alkoxy, alkylthio, cyano, alkoxyalkyloxy and alkylamino groups and halogen atoms), an alkenyl group which may be substituted with halogen atoms, an alkynyl group which may be substituted with halogen atoms, a phosphoric acid ester radical, a cyano group, a group of the following general formula (II):



(wherein  $R^5$  represents an alkyl or alkylamino group which may be substituted with halogen atoms) or a group represented by the following general formula:  $-(R^6)_n - \text{CO} - R^7$  (wherein  $n$  is 0 or 1;  $R^6$  represents a methylene group which may be substituted with alkyl groups, or an ethylene group which may be substituted with alkyl groups;  $R^7$  represents an alkyl group which may be substituted with halogen atoms, an alkoxy group which may be substituted with halogen atoms, an alkylthio group, an alkylamino group or a hydrogen atom);  $Z$  represents a group represented by the general formula:  $\text{N} - \text{R}^8$  ( $R^8$  represents a nitro group, a cyano group, an alkylsulfonyl group which may be substituted with halogen atoms, a tosyl group or an alkylcarbonyl group which may be substituted with halogen atoms) or a group represented by the general formula:  $\text{C}(\text{CN})\text{R}^9$  (wherein  $R^9$  represents a cyano group or an alkoxy carbonyl group); and  $A$  represents an ethylene group which may be substituted with  $C_1$  to  $C_3$  alkyl groups, a trimethylene group which may be substituted with  $C_1$  to  $C_3$  alkyl groups or a group represented by the general formula:  $-\text{CH}_2\text{NR}^{10}\text{CH}_2-$  (wherein  $R^{10}$  is a  $C_1$  to  $C_3$  alkyl group), with a phosphoric acid compound represented by the following general formula (VIII):



(wherein  $R^2$  represents a  $C_1$  to  $C_4$  alkyl group and Hal represents a halogen atom) to obtain a compound represented by the following general formula (IX):



(wherein  $R^2$ ,  $X$ ,  $Z$ ,  $A$  and Hal are the same as those defined above), and further reacting the compound (IX) with a compound represented by the general formula (X):  $\text{R}^1 - \text{OH}$  (wherein  $R^1$  is a  $C^1$  to  $C^4$  alkyl group).

16. An insecticidal, acaricidal or nematocidal composition comprising an appropriate carrier and, as an active ingredient, an organophosphorus compound represented by the following general formula (I) according to any one of claims 1 to 12.



European  
Patent Office

## EUROPEAN SEARCH REPORT

Application Number

EP 91 11 1152

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	EP-A-0 277 317 (NIHON TOKUSHU NOYAKU SEIZO K.K.) * Page 3; claims 1,2,3 * - - - -	1,16	C 07 F 9/6506 A 01 N 57/16 C 07 F 9/6539
Y	EP-A-0 175 985 (SHIONOGI & CO.) * The whole document * - - - -	1,16	C 07 F 9/6521 C 07 F 9/653 C 07 F 9/59
Y	EP-A-0 192 060 (NIHON TOKUSHU NOYAKU SEIZO K.K.) * Compound 453, page 159, claims * - - - - -	1,16	C 07 F 9/572 C 07 F 9/6512 C 07 F 9/6544 C 07 F 9/6533
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C 07 F 9/00 A 01 N 57/00
The present search report has been drawn up for all claims			
Place of search The Hague		Date of completion of search 02 October 91	Examiner OUSSET J-B.
<div>CATEGORY OF CITED DOCUMENTS</div> <div>X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention</div> <div>E: earlier patent document, but published on, or after the filing date D: document cited in the application L: document cited for other reasons &amp;: member of the same patent family, corresponding document</div>			